# Pressure dependence of resistivity and magnetoresistance in Pr-doped La0.7Ca0.3MnO3

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# Pressure dependence of resistivity and magnetoresistance in Pr-doped $La_{0.7}Ca_{0.3}MnO_3$

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We report the effects of magnetic field ($\mu_0 H = 0$–7 T) and hydrostatic pressure ($P = 1$ bar to 9.53 kbar) on electrical resistivity in the phase separated manganite $La_{0.3}Pr_{0.4}Ca_{0.3}MnO_3$. The resistivity shows a first-order transition from paramagnetic insulating to ferromagnetic metallic state at a temperature $T = T_{IM}$ in ambient pressure and zero magnetic field. The first-order transition becomes second-order with increasing pressure and/or magnetic field. Both hydrostatic pressure and magnetic field decrease the magnitude resistivity and shift the resistivity peak at $T = T_{IM}$ towards high temperature with different rates ($dT_{IM}/dH = 13.5$ K/T for P = 1 bar, 8.8 K/T for $P = 9.53$ kbar, and $dT_{IM}/dP \sim 4.42$ K/kbar in zero field). However, the magnitude of the magnetoresistance decreases with increasing pressure. Baroresistance in the absence of magnetic field for $\Delta P = 9.53$ kbar reaches nearly 100% around 150 K. Interestingly, while the resistivity at a constant temperature shows irreversible behaviour upon cycling the direction of magnetic field at ambient pressure, the irreversibility is eliminated under hydrostatic pressure. Our results indicate that pressure eliminates phase separation by converting the paramagnetic polaronic phase into ferromagnetic metallic phase in the vicinity of phase coexistence. 
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Mixed valent manganites have the general formula $R_{1-x}A_xMnO_3$, where R and A are trivalent rare earth and divalent alkaline earth ions. The dramatic decrease of electrical resistance under an external magnetic field shown by these oxides and the physics behind this so called colossal magnetoresistance (CMR) phenomenon aroused enormous attention during the past two decades. Magnetic and electrical properties of these compounds are sensitively dependent on the doping level (x) and the average ionic radius of the R and A cations. For example, for a fixed hole content (x), the low temperature ground state changes from ferromagnetic metal (x = 0) to charge-ordered antiferromagnetic insulator (x = 1) in the series $La_{0.7-x}Pr_xCa_{0.3}MnO_3$.[1] These changes are due to the decrease in Mn-O-Mn bond angle, which controls the one-electron bandwidth as the bigger ionic radius $La^{3+}$ is replaced by smaller ionic radius $Pr^{3+}$. This is called as chemical or internal pressure effect. The ferromagnetic Curie temperature in $R_{0.7}A_{0.3}MnO_3$ series decreases with increasing chemical pressure.[1,2] On the other hand, application of hydrostatic pressure increases the ferromagnetic Curie temperature and decreases the resistivity.[3,4] Although magnetic field and hydrostatic pressure apparently have similar effects on the resistivity, fundamental mechanisms involved are different. In the framework of the double exchange model, the low temperature transport properties are strongly dependent on three parameters, the Mn-O-Mn bond angle ($\phi$), the Mn-O bond lengths ($d_{Mn\text{-}O}$), and the relative angle ($\theta_{ij}$) between Mn spins at the $i_{th}$ and $j_{th}$. The empirical relation for $e_g$-electron bandwidth ($W$) is[5] $W \propto (\cos^2\phi \cos(\theta_{ij}/2))/d^{3.5}_{Mn-O}$.

The effect of magnetic field is primarily to increase spin alignment, causing $\theta_{ij}$ to become very small. On the other hand, structural studies under hydrostatic pressure[6] show that pressure compresses the bond lengths (decreasing $d_{Mn\text{-}O}$) and straightens the Mn-O-Mn bonds (increasing $\phi$ towards 180°). The former augments the overlap of the $e_g$ orbitals of neighboring Mn ions, whereas the latter enhances the oxygen p-orbital based double-exchange coupling. Hence, it will be interesting to study the effect of simultaneous application of both magnetic field and pressure, as it might help us to understand the details of spin-charge-lattice coupling.

Although effects of magnetic field and pressure on resistivity have been reported separately,[7–12] simultaneous actions of magnetic field and pressure on electrical transport are seldom reported.[13] In this work, we report combined effects of pressure and magnetic field on the electrical resistivity in $La_{0.3}Pr_{0.4}Ca_{0.3}MnO_3$ (LPCMO).

The polycrystalline LPCMO sample was prepared by solid-state reaction and was confirmed to be single-phased, from its XRD analysis. Previously, our lab reported magnetic and magnetocaloric studies in $La_{0.7-x}Pr_xCa_{0.3}MnO_3$ series.[14] The pressure studies were done using a piston type Cu-Be pressure cell (Pcell-30 form the easyLab, UK), using 1:1 pentane-isopentane mixture as the transmitting medium. The pressure was calibrated from the superconducting transition of Sn. The resistivity measurements were performed at 1.68, 6.66, and 9.53 kbar, using the AC-transport module in Physical Property Measuring System (PPMS-Quantum

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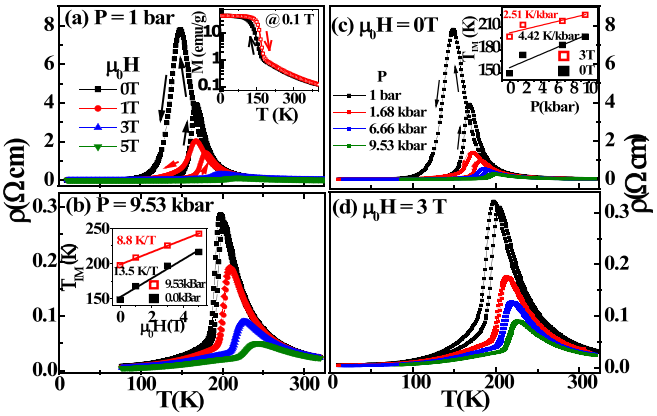


FIG. 1. Temperature dependence of the resistivity under different magnetic fields ($H$) for (a) P = 1 bar, (b) P = 9.53 kbar, and under different pressures (P) for (c) $\mu_0 H = 0$ T and (d) $\mu_0 H = 3$ T. The inset in (a) shows magnetization measured under $\mu_0 H = 1$ kOe. The inset in (b) shows the dependence of the insulator-metal transition temperature ($T_{IM}$) on magnetic field under P = 1 bar and 9.53 kbar. The inset in (d) compares the shift of $T_{IM}$ with pressure under $\mu_0 H = 0$ and 3 T.

Design Inc., USA) with 1 mA ac current of frequency 333 Hz.

Figure 1(a) shows the temperature dependence of the resistivity $\rho(T)$ for the LPCMO sample under different magnetic fields ($\mu_0 H = 0$, 1, 3, 5 T) and at ambient pressure ($P = 1$ bar). Upon cooling in zero magnetic field, $\rho(T)$ shows an insulator to metal transition with a peak in resistivity ($\rho_p$) occurring at $T = T_{IM} = 149.4$ K. This change in electrical behavior is driven by paramagnetic to ferromagnetic transition of $t_{2g}^3$ spins at the $Mn^{3+}$ and $Mn^{4+}$ sites as indicated by the magnetization data (shown in the inset). No evidence of charge ordering in the paramagnetic state is seen in the magnetization data. Upon warming from the low temperature ferromagnetic state, $\rho_p$ shifts to $T_{IM} = 169.65$ K and the peak value of the resistivity is reduced by a factor of 2.5. This large hysteresis behavior in the resistivity is the consequence of the first-order paramagnetic to ferromagnetic transition. In this hysteresis region, both the high temperature paramagnetic phase and low temperature ferromagnetic phase coexist. With increasing $\mu_0 H$, the value of $\rho_p$ plummets rapidly accompanied by an increase in $T_{IM}$ and decrease in hysteresis. $T_{IM}$ increases linearly with $\mu_0 H$ at a rate $dT_{IM}/dH \sim 13.5$ K/T. Fig. 1(b) shows the $\rho(T)$ under different magnetic fields but under the hydrostatic pressure $P = 9.53$ kbar. First, we note that the width of hysteresis in $\rho(T)$ under $\mu_0 H = 0$ T is greatly reduced as compared to the one in ambient pressure data in Fig. 1(a). Second, the magnitude of $\rho_p$ is also reduced and third, the $T_{IM}$ has shifted to higher temperatures, compared to the ambient pressure data. With increasing strength of the external field, $\rho_p$ further reduces in magnitude and shifts up in temperature as in the ambient pressure data. The inset of Fig. 1(b) compares the rate of change of $T_{IM}$ under $P = 1$ bar and $P = 9.53$ kbar; while $dT_{IM}/dH = 13.5$ K/T for $P = 1$ bar, it reduces to 8.8 K/T (smaller by 35%) for P = 9.53 kbar. Fig. 1(c) shows $\rho(T)$ under different hydrostatic pressures ($P = 1$ bar-9.53 kbar) in zero external field. With increasing $P$, the resistivity peaks $\rho_p$ decrease and the $T_{IM}$ shift to high temperatures as in the case of increasing $\mu_0 H$, at ambient pressure in Fig. 1(a). Fig. 1(d) shows $\rho(T)$ under $\mu_0 H = 3$ T for different pressures. The figure is self-explanatory. In the inset of Fig. 1(c), we compare the pressure dependence of $T_{IM}$ for $\mu_0 H = 0$ and 3 T. In zero field, the $T_{IM}$ increases at the rate $dT_{IM}/dP \sim 4.42$ K/kbar but at a smaller rate ($dT_{IM}/dP \sim 2.51$ K/kbar) in the presence of $\mu_0 H = 3$ T. Note that $dT_{IM}/dP$ in zero magnetic field is lower than $dT_{IM}/dH$ (=13.5 K/T) in ambient pressure, the magnetic field is more effective in shifting $T_{IM}$ than hydrostatic pressure.

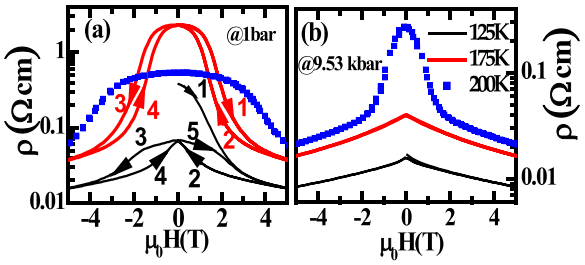


FIG. 2. Magnetic field dependence of the resistivity at selected temperatures for P = 1 bar (a) and 9.53 kbar (b).

Next, we highlight a crucial observation of the present work. We compare the field dependences of the resistivity, $\rho(H)$ at selected temperatures for (a) $P = 1$ bar and (b) $P = 9.53$ kbar in Figs. 2(a) and 2(b), respectively. At T = 200 K under P = 1 bar, $\rho(H)$ varies little with the magnetic field up to H = 2.5 T and thereupon shows a tendency to decrease rapidly. Contrarily, the resistivity under $P = 9.53$ kbar shows a rapid decrease between 0 and 2 T followed by a gradual decrease at higher fields. When $T = 175$ K, $\rho$(H) for $P = 1$ bar varies little with the magnetic field for $\mu_0 H < 1$ T but decreases rapidly between 1 T $< \mu_0 H \leq 3$ T. Upon decreasing the field, $\rho(H)$ shows a clear hysteresis, which is also mirrored in the reverse field sweep. The direction of the field sweeps is sequentially marked in the graph. This hysteresis behavior is completely eliminated under $P = 9.53$ kbar and the $\rho(H)$ decreases gradually with the field starting from $\mu_0 H = 0$ T. The most interesting behavior is seen at 125 K. When $P = 1$ bar, $\rho(H)$ gradually decreases as the magnetic field increases from 0 T to 5 T (marked as path 1) and upon decreasing the magnetic field from 5 T, the $\rho(H)$ takes different path, resulting in irreversible behavior (path 2) at zero field. Upon sweeping the field in the negative direction and reversing again in the positive direction, we observe a reversible hysteresis behavior. On the other hand, the irreversibility is completely eliminated under $P = 9.53$ kbar and the hysteresis is also negligible between the forward and reverse directions of the magnetic field after the initial sweep.

We show the baroresistance, $BR \equiv -[\rho(P = 0, \mu_0 H) - \rho(P = 9.53\ \text{kbar}, \mu_0 H)]/\rho(P = 0, \mu_0 H)$, calculated for different magnetic fields for a fixed pressure difference ($\Delta P = 9.53$ kbar) in Fig. 3(a). The BR in zero field increases gradually as the temperature decreases from 300 K. It shows a rapid increase around 200 K, goes through a broad maximum around 175 K, where it reaches ~100% and then decreases. As the field increases, the broad maximum evolves into a clear peak, decreases in magnitude, and shifts up in temperature. In a field

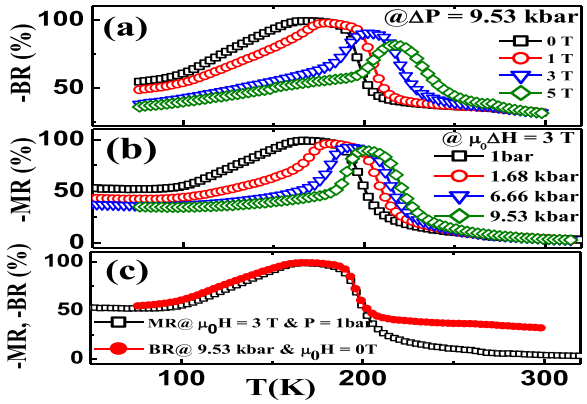


FIG. 3. (a) Baroresistance (BR) as a function of temperature at different magnetic fields ($H$) for a pressure difference of $\Delta P = 9.53$ kbar. (b) MR as a function of temperature for different pressures (P) for a fixed magnetic field difference ($\mu_0 \Delta H = 3$ T). (c) A comparison of BR for P = 9.53 kbar at $\mu_0 H = 0$ T and MR for $\mu_0 H = 3$ T at P = 1 bar.

of $\mu_0 H = 3$ T, the BR has decreased to ~90%. Fig. 3(b) shows the magnetoresistance, $MR = -[\rho(P, H = 0) - \rho(P, H = 3\,T)]/\rho(P, H = 0))$ calculated for different pressures but for a fixed magnetic field difference ($\mu_0 \Delta H = 3$ T). In ambient pressure, the MR reaches a maximum value of 98% around 150 K. With increasing pressure, the MR peak decreases and shifts up in temperature. The MR decreases to 90% for $P = 9.53$ kbar. For comparison of the pressure induced and magnetic field induced resistance changes, we plot MR for $\mu_0 H = 3$ Tesla measured at ambient pressure (P = 1 bar) and BR for P = 9.53 kbar measured at $\mu_0$H = 0 T in Fig. 3(c). The difference between BR and MR is small in the ferromagnetic state. It is clear that BR in the paramagnetic state (>200 K) is far greater than the MR. For example, at T = 250 K, the MR is only 9.2% but the BR reaches 37.4%.

The salient features of the presented results can be summarized as follows: (1) Both the pressure and the magnetic field decrease the resistivity peak and shift the insulator-metal transition temperature ($T_{IM}$) towards high temperature side. However, the rate of increase of $T_{IM}$ with $\mu_0 H$ is higher than with P. (2) Pressure clearly eliminates the irreversibility in the magnetic field dependence of the resistivity at temperature below the $T_{IM}$. The field dependence is clearly modified: the resistance changes more rapidly at lower magnetic fields under pressure compared to the case of ambient pressure. (3) Baroresistance above $T_C$ is much larger than the magnetoresistance, while it is comparable in the ferromagnetic state. The last point suggests that the high sensitivity of the charge carriers of the paramagnetic phase to the hydrostatic pressure and similar effect was observed earlier in other manganites in the absence of magnetic field. It is known that dynamic Jahn-Teller distortions enhance electron-phonon coupling which localize the charge carriers as small polarons in the paramagnetic phase. Electrical conduction occurs due to hopping by these small polarons among the neighboring sites. Application of the hydrostatic pressure weakens the electron-phonon coupling and increases the polaron mobility. As $T_C$ is approached from the high temperature side, polarons concentration increase and they merge around $T_C$. The resistivity exhibits a pronounced hysteresis around $T_{IM}$ as the high temperature paramagnetic polaronic insulating and the low temperature ferromagnetic metallic phase coexist in this first-order transition. These two phases also have different structural parameters because charge carriers are localized as polarons in the paramagnetic phase but are itinerant in the ferromagnetic phase.[14,15] The interfacial strain between these two mixed phases prevents complete transformation of the paramagnetic phase into ferromagnetic phase unless thermal energy exceeds the interfacial energy. The irreversibility observed at 125 K in the ambient pressure is the consequence of the phase separation. As the field initially increases from 0 T to the maximum field, apart from ferromagnetic domains rotating towards the direction of the magnetic field, paramagnetic regions in the proximity of ferromagnetic domains also transform into ferromagnetic phase. It needs more magnetic energy to move the phase boundary between the paramagnetic and ferromagnetic phases. However, once the sample becomes magnetically homogeneous at a high field, it does not revert back to the initial zero-field domain configuration, resulting in the observed irreversibility. Pressure compresses the Mn-O bond length, which increases the overlap of Mn-$e_g$–O-2p orbitals and hence enhances mobility of $e_g$ holes which in turn enhances the double-exchange interaction in the paramagnetic phase. Thus, the application of pressure seems to convert the paramagnetic phase into ferromagnetic phase easily than the magnetic field, i.e., it enhances the volume fraction of the ferromagnetic phase. As a result, the field dependence of the resistivity under a high pressure shows negligible irreversibility.

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